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PAGE 1/8 \* RCVD AT 8/14/2007 5:39:59 PM [Eastern Daylight Time] \* SVR:USPTO-EFXRF-6/22 \* DNI:2738300 \* CSID:7032041352 \* DURATION (mm:ss):01:50

AUG 14 2007

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re U.S. Patent Application of )  
 YAMAZAKI et al. )  
 Application Number: 10/576,677 )  
 Filed: April 21, 2006 )  
 For: MULTILAYER FILM )  
 Attorney Docket No. NISH.0003 )

Art Unit 1772

Fax 571-273-8300

**Commissioner for Patents**  
**P.O. Box 1450**  
**Alexandria, VA 22313-1450**

LETTER

Sir:

The below-identified communications are submitted in the above-captioned application or proceeding:

(x) Request for Corrected Publication under 37.C.F.R. §1.221(b)  
 (x) US 2007/0134476 A1 pages 5, 9 and 12

The Commissioner is hereby authorized to charge payment of any fees associated with this communication, including fees under 37 C.F.R. § 1.16 and 1.17 or credit any overpayment to Deposit Account Number 08-1480. A duplicate copy of this sheet is attached.

Respectfully submitted,

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 August 14, 2007

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Art Unit 1772

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Commissioner of Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450REQUEST FOR CORRECTED PUBLICATION UNDER 37 C.F.R. §1.221(b)

Applicants respectfully request that the published application in connection with the above-referenced application be corrected in accordance with the attached marked-up copy of the published application. Applicants believe that correction is proper on the grounds that a material mistake by the Office was made since the published application failed to include a claim that was part of the original application as filed and was not canceled by preliminary amendment.

Specifically, upon a review of the Notice of Publication of Application dated June 14, 2007, Applicants noted that claim 11, line 12 of the published application read in the relevant part "... with ASTM D 3985-81 is retained within 50% of . . ." However, as originally filed, claim 11 read in the relevant part "... with ASTM D 3985-81 is retained within ±50% of . . ." In addition, Applicants noted that the paragraphs were numbered incorrectly starting at paragraph [0052] where the subtitle "3. Production Process of Multi-Layer Film:" and the subtitle "(4) Measurement of Concentration Gradient Structure" were incorrectly numbered as paragraphs, which was inconsistent with other similar subtitles that were not numbered.

Attached hereto are the pages from the above-referenced published application with the errors as outlined above have been marked.

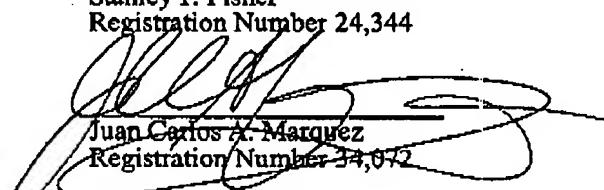
In accordance with the requirements of MPEP §1130, Applicants have checked PAIR, and have confirmed that the application documents as originally filed did not contain the errors for which correction is requested and that the text was legible.

The Notice of Publication was dated June 14, 2007, the period for response to which is set to expire on August 14, 2007. Therefore, this request is timely, and Applicants respectfully requests that the error be corrected and a corrected publication be issued.

If there are any fees that may be due or be required by the submission of this request, the Commissioner is authorized to credit any overpayment and charge any deficiency in any fees required under 37 CFR §§ 1.16, 1.17 and/or 1.18 to Deposit Account No. 08-1480.

Please direct any inquiries in connection with this application to the Applicants' undersigned representative.

Respectfully submitted,

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## [0052] 3. Production Process of Multi-Layer Film:

[0053] The multi-layer film according to the present invention can be produced by a process, in which at least one carboxyl group-containing polymer layer and at least one polyvalent metal compound-containing layer are formed adjacently to each other on a base (support) by a coating method. The polyvalent metal compound migrates from the polyvalent metal compound-containing layer into the carboxyl group-containing polymer layer to form a polyvalent metal salt with the carboxyl group in the carboxyl group-containing polymer there, thereby introducing an ionic bond (i.e., ionic crosslinking).

[0054] Although it is necessary for the carboxyl group-containing polymer layer to adjoin the polyvalent metal compound-containing layer, the number of the respective layers arranged may be suitably determined as needed. When the carboxyl group-containing polymer layer and the polyvalent metal compound-containing layer are indicated as A and B, respectively, specific examples of multi-layer structures include A/B, B/A/B and A/B/A. Of course, multi-layer structures having at least 4 layers, such as A/B/A/B, B/A/B/A and B/A/B/A/B, may also be adopted.

[0055] As the base, is preferably used a plastic film. No particular limitation is imposed on the kind of a plastic forming the plastic film. As examples thereof, may be mentioned olefin polymers such as high density polyethylene, medium density polyethylene, low density polyethylene, linear low density polyethylene, polypropylene, poly(4-methylpentene) and cyclic polyolefins, and acid-modified products thereof; vinyl acetate polymers such as polyvinyl acetate, ethylene-vinyl acetate copolymers, esterified products of ethylene-vinyl acetate copolymers and polyvinyl alcohol, and modified products thereof; polyesters such as polyethylene terephthalate, polybutylene terephthalate and polyethylene naphthalate; aliphatic polyesters such as poly( $\epsilon$ -caprolactone), polyhydroxybutyrate and polyhydroxyvalerate; polyamides such as nylon 6, nylon 66, nylon 12, nylon 6/66 copolymers, nylon 6/12 copolymers and meta-xylenediparaffide-nylon 6 copolymers; polyethers such as polyethylene glycol, poly(ether sulfone), poly(phenylene sulfide) and poly(phenylene oxide); halogenated polymers such as polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride and polyvinylidene fluoride; acrylic polymers such as polymethyl acrylate, polyethyl acrylate, polymethyl methacrylate, polyethyl methacrylate and polyacrylonitrile; polyimide resins; and besides resins used for paints, such as alkyl resins, melamine resins, acrylic resins, pyroxylin, urethane resins, unsaturated polyester resins, phenol resins, amino resins, fluorocarbon resins and epoxy resins; and natural polymeric compounds such as cellulose, starch, pullulan, chitin, chitosan, glucosmannan, agarose and gelatin.

[0056] The base is preferably an uncoated film or oriented film formed of any one of these plastics. As the base, may also be used that obtained by forming a thin film of an inorganic compound such as silicon oxide, aluminum oxide, aluminum or silicon nitride; a metal compound; or the like on the surface of the film (including a sheet) formed of the plastic by a vapor deposition method sputtering method or ion plating method.

[0057] In order to form the carboxyl group-containing polymer layer, a coating liquid comprising the carboxyl group-containing polymer and a solvent is prepared, and this

coating liquid is applied on to the base or the polyvalent metal compound-containing layer and dried. No particular limitation is imposed on the solvent so far as it can uniformly dissolve or disperse the carboxyl group-containing polymer therein. As specific examples thereof, may be mentioned water, alcohols such as methyl alcohol, ethyl alcohol and isopropyl alcohol; and polar organic solvents such as dimethyl sulfoxide, N,N-dimethylformamide and N,N-dimethylacetamide.

[0058] No particular limitation is imposed on the concentration of the carboxyl group-containing polymer in the coating liquid comprising the carboxyl group-containing polymer and the solvent. However, it is desirable that the concentration be within a range of preferably 0.1 to 50% by weight, more preferably 1 to 40% by weight, particularly preferably 5 to 30% by weight from the viewpoints of the stability, uniformly-coating ability and easy coating operation of the coating liquid. Into the coating liquid comprising the carboxyl group-containing polymer, additive components such as other polymers, softening agents, stabilizers, anti-blocking agents, pressure-sensitive adhesives, inorganic lamellar compounds typified by montmorillonite, colorants and ultraviolet absorbents may be suitably added within limits not impeding the gas barrier property as needed. It is desirable that the amount added be preferably at most 5% by weight, more preferably at most 3% by weight, particularly preferably at most 1% by weight in terms of the total amount of the additives based on the weight of the carboxyl group-containing polymer.

[0059] The polyvalent metal compound-containing layer can be formed by applying a coating liquid comprising the polyvalent metal compound on to the base or the carboxyl group-containing polymer layer and drying it. The coating liquid comprising the polyvalent metal compound can be prepared by dissolving or dispersing the polyvalent metal compound in a solvent.

[0060] No particular limitation is imposed on the solvent so far as it can uniformly dissolve or disperse the polyvalent metal compound therein. As specific examples thereof, may be mentioned water, alcohols such as methyl alcohol, ethyl alcohol, isopropyl alcohol, *n*-propyl alcohol, *n*-butyl alcohol and *n*-pentyl alcohol; ketones such as acetone and methyl ethyl ketone; ethers such as diethyl ether, dioxane and tetrahydrofuran; esters such as ethyl acetate and butyl acetate; polar organic solvents such as dimethyl sulfoxide, N,N-dimethylacetamide, N,N-dimethylformamide, N-methyl-2-pyrrolidone, tetramethylurea, hexamethylphosphoric triamide and  $\gamma$ -butyrolactone; and hydrocarbons such as toluene, hexane, heptane and cyclohexane. These solvents may be used either singly or in any combination thereof.

[0061] Into the coating liquid comprising the polyvalent metal compound, resin components and additives such as dispersing agents, surfactants, softening agents, stabilizers, film-forming agents, anti-blocking agents and pressure-sensitive adhesives may be suitably added as needed. A resin component soluble in the solvent used is preferably contained from the viewpoint of improving the dispersibility of the polyvalent metal compound and the coating property of the coating liquid.

[0062] As examples of the resin component added to the coating liquid comprising the polyvalent metal compound, may be mentioned general-purpose binder resins for paints,

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of  $1700\text{ cm}^{-1}$  assigned to a carboxyl group ( $-\text{COOH}$ ). In other words, the  $\text{C}=\text{O}$  stretching vibration assigned to the carboxyl group ( $-\text{COOH}$ ) gives an absorption peak having an absorption maximum in the vicinity of  $1700\text{ cm}^{-1}$  in an infrared ray wave number region of from  $1800\text{ cm}^{-1}$  to  $1600\text{ cm}^{-1}$ .

[0092] The absorbance of a film has a proportional relation to an amount of a chemical species having infrared activity, which is present in the film. Accordingly, the peak ratio  $A_{1600}/A_{1700}$  of the infrared absorption spectrum may be used as an index indicating a quantitative ratio of the carboxylate ( $-\text{COO}^-$ ) with the carboxyl group of the carboxyl group-containing polymer formed with the polyvalent metal in the film to the free carboxyl group ( $-\text{COOH}$ ). This ratio is also an index indicating the degree of ionization.

#### (2) Oxygen Transmission Rate:

[0093] An oxygen transmission rate of a film was measured under conditions of a temperature of  $30^\circ\text{ C}$ . and a relative humidity of 0% by means of an oxygen transmission tester, Oxtran 2/20 manufactured by Modern Control Co. The measurement was carried out in accordance with ASTM D 3985-B1 (corresponding to the B method of JIS K 7126). The unit of the measured value is  $\text{cm}^3(\text{STP})/(\text{m}^2 \text{ day-MPa})$ . "STP" means standard conditions ( $0^\circ\text{ C}$ , 1 atm) for defining the volume of oxygen.

[0094] The oxygen transmission rate was measured before and after a flex test, which will be described subsequently. After the flex test, a sample was aged for 20 hours under an environment of a temperature of  $30^\circ\text{ C}$ . and a relative humidity of 80%. With respect to this aged sample (humidity conditioned sample), the oxygen transmission rate was measured likewise.

#### (3) Flex Test:

[0095] A flex test was carried out by means of a Gelvo tester under conditions of a temperature of  $5^\circ\text{ C}$ . and a relative humidity of 10% in accordance with the provisions of ASTM F 392. The test is a method that a process of twisting a film formed into a cylindrical form and further compressing it is repeated, and was used as a testing method for resistance to flex fatigue. Specifically, a multi-layer film formed into a cylindrical form was used as a sample to conduct a flex test of 150 Gelvo flexings by means of a Gelvo tester under the above-described conditions.

#### (4) Measurement of Concentration Gradient Structure:

[0097] A multi-layer film intended to be observed was embedded in an epoxy resin (product of NISSHIN-BM CORPORATION, trade name "Quentol 812"). This embedded sample was trimmed into an about 0.5-by-0.5 mm square to obtain an ultrathin section by means of an ultramicrotome (manufactured by Reichert-Nisai Co., trade name "ULTRACUT. N"). The thickness of this ultrathin section was about 100 nm.

[0098] Image observation of the thus-obtained ultrathin section through an transmission electron microscope (TEM)

was conducted by means of a transmission electron microscope, HF-2000 manufactured by Hitachi Ltd. to determine measuring points. An elemental composition distribution in the thickness-wise direction of a section of the film was measured by energy dispersive X-ray spectroscopy (EDX). Measuring conditions of the energy dispersive X-ray spectroscopy are as follows.

[0099] Measuring apparatus: elemental analyzer, VOYAGER II manufactured by NORAN Co.,

[0100] X-ray detector: Si/Li semiconductor detector,

[0101] Energy resolution: 137 eV,

[0102] X-ray take-off angle:  $22^\circ$  (side take up system),

[0103] Acceleration voltage: 200 kV,

[0104] Beam diameter: about 1 nm, and

[0105] Incorporation time: for 30 seconds.

[0106] The concentration distribution of a polyvalent metal element component in the polymer layer containing the carboxyl group-containing polymer and the polyvalent metal salt of the carboxyl group-containing polymer was evaluated by a ratio M/C of the number M of polyvalent metal atoms to the number C of carbon atoms.

[0107] The polyvalent metal element in the polymer layer forms a salt with the carboxyl group, and the concentration distribution of a polyvalent metal element in the thickness-wise direction indicates a concentration distribution of the polyvalent metal salt of the carboxyl group-containing polymer in the polymer layer. Namely, the ratio M/C of the number M of polyvalent metal atoms to the number C of carbon atoms was used for identifying the gradient structure.

#### Example 1

[0108] An aqueous solution of polyacrylic acid (product of Togosei Chemical Industry Co., Ltd., trade name "Aron™ A-10H", concentration: 25% by weight, number average molecular weight: 200,000) was diluted with distilled water to prepare an aqueous solution of polyacrylic acid (PAA) having a concentration of 10% by weight. The thus-obtained aqueous solution was applied on to an oriented polyethylene terephthalate film (PET film, product of Toray Industries, Inc., trade name "Lumirror™ S10", thickness: 12  $\mu\text{m}$ , heat shrink: 0.5% as measured by dipping for 30 seconds in hot water of  $90^\circ\text{ C}$ .) by means of a bar coater (manufactured by RK PRINT-COAT INSTRUMENT Co., trade name "K303 PROOFERT™"), and a coating film was dried by a dryer. The dry coating weight of polyacrylic acid was  $0.90\text{ g/m}^2$ , and the dry thickness of the coating film was 1.0  $\mu\text{m}$ .

[0109] The thus-obtained PAA film was coated with an ultrafine particulate zinc oxide-containing paint (product of Sumitomo Osaka Cement Co., Ltd., trade name "ZR133", total nonvolatile component: 33% by weight, zinc oxide ultrafine particles: 18% by weight) by means of the same bar coater as described above, and a coating film was dried to produce a multi-layer film having a layer structure of "PET/PAA/ZnO-containing layer". The dry coating weight

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layer (A) in the thickness-wise direction from a surface adjoining each of the polyvalent metal compound-containing layers (B).

5. The multi-layer film according to claim 1, wherein the concentration gradient structure of the polymer layer (A) has a low concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of 0 to 0.06/n, in a proportion of 5 to 80% in terms of a thickness percentage of the polymer layer (A).

6. The multi-layer film according to claim 1, wherein the concentration gradient structure of the polymer layer (A) has a high concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of from higher than 0.06/n to not higher than 0.33/n, in a proportion of 20 to 95% in terms of a thickness percentage of the polymer layer (A) adjoining the polyvalent metal compound-containing layer (B).

7. The multi-layer film according to claim 2, wherein the polymer layer (A) has a high concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of from higher than 0.06/n to not higher than 0.33/n, in a proportion of 20 to 95% in terms of a thickness percentage of the polymer layer (A) adjoining the polyvalent metal compound-containing layer (B), and has, on the side opposite to the surface adjoining the polyvalent metal compound-containing layer (B), a low concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] falls within a range of 0 to 0.06/n, in a proportion of 5 to 80% in terms of a thickness percentage of the polymer layer (A).

8. The multi-layer film according to claim 3, wherein each of the polymer layers (A) has a high concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of from higher than 0.06/n to not higher than 0.33/n, in a proportion of 20 to 95% in terms of a thickness percentage of the polymer layer (A) adjoining the polyvalent metal compound-containing layer (B), and has, on the side opposite to the surface adjoining the polyvalent metal compound-containing layer (B), a low concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] falls within a range of 0 to 0.06/n, in a proportion of 5 to 80% in terms of a thickness percentage of the polymer layer (A).

9. The multi-layer film according to claim 4, wherein the polymer layer (A) has a high concentration region of the polyvalent metal salt of the carboxyl group-containing poly-

mer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of from higher than 0.06/n to not higher than 0.33/n, in a proportion of 20 to 95% in terms of a thickness percentage of the polymer layer (A) adjoining each of the polyvalent metal compound-containing layers (B), and has, at a central portion thereof, a low concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] falls within a range of 0 to 0.06/n, in a proportion of 5 to 80% in terms of a thickness percentage of the polymer layer (A).

10. The multi-layer film according to claim 1, wherein a chemical equivalent of the polyvalent metal compound to the carboxyl group, which is calculated out on the basis of the total (At) of the carboxyl group contained in the whole carboxyl group-containing polymer layer (A) and the total (Bt) of the polyvalent metal compound contained in the whole carboxyl group-containing polymer layer (A) and the whole polyvalent metal compound-containing layer (B), is at least 1.0.

11. The multi-layer film according to claim 1, wherein an oxygen transmission rate as determined by using the multi-layer film formed into a cylindrical form as a sample to conduct a flex test of 150 Gelvo flexings by means of a Gelvo tester under conditions of a temperature of 5° C. and a relative humidity of 10% in accordance with the provisions of ASTM F 392, aging the sample for 20 hours under an environment of a temperature of 30° C. and a relative humidity of 80%, and then measuring an oxygen transmission rate of the sample after the aging under conditions of a temperature of 30° C. and a relative humidity of 80% in accordance with ASTM D 3985-81 is retained within 50% of the oxygen transmission rate of the sample before the flex test.

12. The multi-layer film according to claim 1, wherein the carboxyl group-containing polymer has an oxygen transmission coefficient of at most 1,000  $\text{cm}^2 \mu\text{m}/(\text{m}^2 \text{day MPa})$  as determined in the form of a film formed by itself under low-humidity conditions of a temperature of 30° C. and a relative humidity of 0%.

13. The multi-layer film according to claim 1, wherein the carboxyl group-containing polymer is a homopolymer of a carboxyl group-containing unsaturated monomer, a copolymer of carboxyl group-containing unsaturated monomer, a copolymer of a carboxyl group-containing unsaturated monomer and any other polymerizable monomer, a carboxyl group-containing polysaccharide or a mixture of at least two monomers thereof.

14. The multi-layer film according to claim 1, wherein the carboxyl group-containing unsaturated monomer is at least one  $\alpha,\beta$ -monoethylenically unsaturated carboxylic acid selected from the group consisting of acrylic acid, methacrylic acid, crotonic acid, itaconic acid, maleic acid and fumaric acid.

15. The multi-layer film according to claim 1, wherein the polyvalent metal compound is an oxide, carbonate, organic acid salt or alkoxide of beryllium, magnesium, calcium, copper, cobalt, nickel, zinc, aluminum or zirconium.

16. The multi-layer film according to claim 1, wherein the polyvalent metal compound is a divalent metal compound.